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Electron Dynamics During High-Power, Short-Pulsed Laser Interactions with Solids and Interfaces

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The objective of the research program is to explore the effects of spatially-confined ultrafast optical excitations of materials to a state of strong electron-phonon nonequilibrium on the evolution of the deposited energy, electronic scattering processes and resulting thermal transport properties. The inadequate physical understanding of the processes that control the temporal and spatial energy confinement in short-pulsed laser interactions with materials inhibits the advancement of laser processing applications. Therefore, the overarching goal of the proposed work is to investigate electronic excitation parameters and the material thermal response to high-power, short-pulse laser excitations at different spatial and temporal scales. In particular, this project investigates the combined effects of laser pulse properties and sample geometry on short-pulse laser processing of nanostructured materials in an effort to control the level of electronic excitation and resulting energy confinement based on laser and interfacial parameters. Using thermoreflectance-based laser techniques to probe samples on which electric fields are applied, this work also demonstrated the ability to continuously tune the phonon thermal conductivity of ferroelectric solids.

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Program: Plasma and Electro-Energetic Physics (RTB-5)
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Abstract (repeated from Abstract field in online form)
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Quantitative Metrics

- **1 postdoctoral research** partially supported
- **1 graduate student** fully supported
- **10 referred journal publications** (listed below and appended to end of report)
- Editor for **2 conference proceeding volumes**
- Work presented as part of **23 contributed conference presentations**
- Work presented by Patrick as part of **5 invited talks**

Published Journal Papers (Full articles appended at end of this report)

1) Q. Zhou, A. Cross, Y. Fu, A. Beling, B. Foley, P. Hopkins, and J. Campbell. Balanced InP/InGaAs photodiodes with 1.5-W output power. Photonics Journal, IEEE, 5(3):6800307, 2013.

2) P. E. Hopkins, C. Adamo, L. Ye, B. D. Huey, S. R. Lee, D. G. Schlom, and J. F. Ihlefeld. Effects of coherent ferroelastic domain walls on the thermal conductivity and kapitza conductance in bismuth ferrite. Applied Physics Letters, 102(12):121903, 2013.

3) R. E. Jones, J. C. Duda, X. W. Zhou, C. J. Kimmer, and P. E. Hopkins. Investigation of size and electronic effects on Kapitza conductance with non-equilibrium molecular dynamics. Applied Physics Letters, 102(18):183119, 2013.

4) C. B. Saltonstall, J. Serrano, P. M. Norris, P. E. Hopkins, and T. E. Beechem. Single element raman thermometry. Review of Scientific Instruments, 84(6):064903, 2013.

5) P. E. Hopkins, J. C. Duda, B. Kaehr, X. Wang Zhou, C.-Y. Peter Yang, and R. E. Jones. Ultrafast and steady-state laser heating effects on electron relaxation and phonon coupling mechanisms in thin gold films. Applied Physics Letters, 103(21):211910, 2013.

6) A. Giri, B. M. Foley, and P. E. Hopkins. Influence of hot electron scattering and electron-phonon interactions on thermal boundary conductance at metal/nonmetal interfaces. Journal of Heat Transfer, 136:092401, 2014.

7) A. Giri, J. T. Gaskins, B. M. Foley, R. Cheaito, and P. E. Hopkins. Experimental evidence of excited electron number density and temperature effects on electron-phonon coupling in gold films. Journal of Applied Physics, 117(4):044305, 2015.

8) A. Giri, J. T. Gaskins, B. F. Donovan, C. Szwejkowski, R. J. Warzoha, M. A. Rodriguez, J. Ihlefeld, and P. E. Hopkins. Mechanisms of nonequilibrium electron-phonon coupling and thermal conductance at interfaces. Journal of Applied Physics, 117(10):105105, 2015.

9) R. Cheaito, K. Hattar, J. T. Gaskins, A. K. Yadav, J. C. Duda, T. E. Beechem, J. F. Ihlefeld, E. S. Piekos, J. K. Baldwin, A. Misra, and P. E. Hopkins. Thermal flux limited

electron kapitza conductance in copper-niobium multilayers. Applied Physics Letters, 106:093114, 2015.

10) J. F. Ihlefeld, B. M. Foley, D. A. Scrymgeour, J. R. Michael, B. B. McKenzie, D. L. Medlin, M. Wallace, S. Trolier-McKinstry, and P. E. Hopkins. Room-temperature voltage tunable phonon thermal conduc- tivity via reconfigurable interfaces in ferroelectric thin films. Nano Letters, 15:1791–1795, 2015.
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Edited Conference Proceedings Volumes
1) **Hopkins, P.E.** (Ed.), *Nanoscale Heat Transport – From Fundamentals to Devices*, in Material Research Society Symposium Proceedings **1779** (Cambridge University Press, MRS Online Proceedings Library Archive, 2015).

2) Beckman, S. P., Bottner, H., Chalopin, Y., Dames, C., Greaney, P.A., **Hopkins, P.E.**, Li, B., Mori, T., Nishimatsu, T., Pipe, K., Venkatasubramanian, R. (Eds.), *Nanoscale Thermoelectric Materials: Thermal and Electrical Transport, and Applications to Solid-State Cooling and Power Generation*, in Material Research Society Symposium Proceedings **1543** (Cambridge University Press, New York, 2013).

Conference Presentations
1) Cheaito, R., Duda, J.C., Beechem, T.E., Medlin, D.L, Hattar, K., Piekos, E.S., Hopkins, P.E., "The effect of ballistic electron transport on copper-niobium thermal interface conductance," 2013 Materials Research Society Spring Meeting, San Francisco, CA, April 1 – 5, 2013 (Poster).

2) Foley, B.M., Kittiwatanakui, S., Duda, J.C., Lu, J., Hopkins, P.E., "Crossover from phonon to electrons dominated thermal boundary conductance of vanadium dioxide thin films across the metal-insulator-transition," 2013 Materials Research Society Spring Meeting, San Francisco, CA, April 1 – 5, 2013.

3) Hopkins, P.E., Ihlefeld, J.F., Foley, B.M., Brown-Shaklee, H.J., Adamo, C., Ye, L., Huey, B., Lee, S., Schlom, D.G., "Strain field and coherent domain wall effects on thermal conductivity and Kapitza conductance across internal boundaries," 2013 Materials Research Society Spring Meeting, San Francisco, CA, April 1 – 5, 2013.

4) Hopkins, P.E., Adamo, C., Ye, L., Huey, B.D., Lee, S.R., Schlom, D.G., Ihlefeld, J.F., "Effects of coherent ferroelastic domain walls on the thermal conductivity and Kapitza conductance in bismuth ferrite," 2013 Summer Heat Transfer Conference, Minneapolis, MN, July 14 – 19, 2013.

5) Foley, B.M., Kittiwatanakul, S., Duda, J.C., Lu, J., Hopkins, P.E., “Crossover from phonon to electron dominated thermal boundary conductance of vanadium dioxide thin films across the metal-insulator-transition,” 2013 Summer Heat Transfer Conference, Minneapolis, MN, July 14 – 19, 2013.

6) . Cheaito, R., Duda, J.C., Beechem, T.E., Hattar, K., Ihlefeld, J.F., Piekos, E., Misra, A., Baldwin, J.K., Hopkins, P.E., “The effect of ballistic electron transport on copper-niobium thermal interface conductance,” 2013 Summer Heat Transfer Conference, Minneapolis, MN, July 14 – 19, 2013.

7) Giri, A., Foley, B.M., Duda, J.C., Hopkins, P.E., “Influence of hot electron scattering on electron-phonon equilibrium in thin film gold systems,” 3rd Annual NanoWorcester Symposium, Worcester, MA, September 28, 2013.

8) Cheaito, R., Gaskins, J.T., Duda, J.C., Beechem, T.E., Ihlefeld, J.F., Hattar, K., Piekos, E.S., Misra, A., Baldwin, J.K., Hopkins, P.E., “The effect of ballistic electron transport on copper-niobium thermal interface,” 3rd Annual NanoWorcester Symposium, Worcester, MA, September 28, 2013.

9) Foley, B.M., Brown-Shaklee, H.J., Donovan, B.F., Duda, J.C., Campion, M.J., Medlin, D.L., Clem, P.G., Ihlefeld, J.F., Hopkins, P.E., “Phonon scattering in nanostructured ferroelectric oxides,” 16th US-Japan Seminar on Dielectric and Piezoelectric Ceramics, Raleigh, NC, November 3 – 6, 2013 (Poster).

10) Giri, A., Hopkins, P.E., “Theory on electron-phonon coupling and thermal conductance at free electron metal/insulator interfaces,” 2013 ASME International Mechanical Engineering Congress & Exposition, San Diego, CA, November 15 – 21, 2013 (Poster).

11) Giri, A., Hopkins, P.E., “Ultrafast and steady-state leaser heating effects on electron relaxation and phonon coupling mechanisms in thin gold films,” 2013 ASME International Mechanical Engineering Congress & Exposition, San Diego, CA, November 15 – 21, 2013.

12) Hopkins, P.E., “Phonon transport in complex oxide nanostructures: grain scattering, domain interactions, and coherent transport,” 32nd International Thermal Conductivity Conference/20th Annual Thermal Expansion Symposium, West Lafayette, IN, April 27 – May 1, 2014.

13) Giri, A., Gaskins, J.T., Hopkins, P.E., “Influence of non-thermal electron dynamics on electron-phonon coupling in thin gold films,” AIAA AVIATION 2014, Atlanta, GA, June 16 – 20, 2014.

14) Hopkins, P.E., Foley, B.M., Cheaito, R., Donovan, B.F., Yadav, A., Rossen, P., Ramesh, R., Majumdar, A., Medlin, D., Brown-Shaklee, H., Ihlefeld, J., Ravichandran, J., “Phonon Transport Processes in Complex Oxide Nanostructures: Coherent

Transport, Grain Scattering, and Domain Interactions," Materials Science & Technology 2014, Pittsburgh, PA, October 12 – 16, 2014.

15) Foley, B.M., Ihlefeld, J.F., Wallace, M., Scrymgeour, D., Michael, J., McKenzie, B., Medlin, D.L., Trolier-McKinstry, S.E., Hopkins, P.E., "Tunable thermal conductivity in a bilayer PZT thin film via applied electric fields," Center for Dielectrics and Piezoelectrics Fall Meeting, Raleigh, NC, November 3 – 4, 2014 (Poster).

16) Hopkins, P.E., Foley, B.M., Cheaito, R., Ravichandran, J., Yadav, A., Rossen, P., Ramesh, R., Majumdar, A., Medlin, D., Brown-Shaklee, H., Ihlefeld, J., "Phonon Transport Processes in Complex Oxide Nanostructures: Coherent Transport, Grain Scattering, and Domain Interactions," 2014 ASME International Mechanical Engineering Congress & Exposition, Montreal, Canada, November 14 – 20, 2014.

17) Giri, A., Gaskins, J.T., Cheaito, R., Foley, B.M., Hopkins, P.E., "Experimental study of electron relaxation and electron-phonon coupling dependence on electron distribution, lattice temperature, substrate, and interface adhesion," 2014 ASME International Mechanical Engineering Congress & Exposition, Montreal, Canada, November 14 – 20, 2014.

18) Foley, B.M., Scrymgeour, D., Medlin, D.L., Ihlefeld, J.F., Hopkins, P.E., "Tunable thermal conductivity in a bilayer PZT thin films via modulation of the domain population distribution using applied electric fields," 2014 Materials Research Society Fall Meeting, Boston, MA, November 30 – December 5, 2014 (Poster).

19) Foley, B.M., Ihlefeld, J., Wallace, M., Scrymgeour, D.A., Michael, J.R., McKenzie, B.B., Medlin, D.L., Trolier-McKinstry, S., Hopkins, P.E., "Tunable thermal conductivity over temperature in bilayer and strain-released PZT thin films via modulation of the domain structure using applied electric fields," Electronic Materials and Applications 2015, Orlando, FL, January 21 – 23, 2015.

20) Ihlefeld, J., Foley, B.M., Scrymgeour, D., Michael, J.R., McKenzie, B.B., Medlin, D.L., Desmarals, J., Wallace, M., Adamo, C., Huey, B.D., Trolier-McKinstry, S., Schlom, D., Hopkins, P.E., "Room temperature voltage tuning of thermal conductivity in ferroelectric thin films," Electronic Materials and Applications 2015, Orlando, FL, January 21 – 23, 2015.

21) Foley, B.M., Ihlefeld, J.F., Wallace, M., Scrymgeour, D., Michael, J., McKenzie, B., Medlin, D.L., Trolier-McKinstry, S.E., Hopkins, P.E., "Tunable thermal conductivity over temperature in bilayer and strain released PZT thin films via modulation of the domain structure using applied electric fields," 2015 Materials Research Society Spring Meeting, San Francisco, CA, April 6 – 10, 2015.

22) Giri, A., Gaskins, J.T., Donovan, B., Szwejkowski, C.J., Warzoha, R., Rodriguez, M., Ihlefeld, J.F., Hopkins, P.E., "Mechanisms of nonequilibrium electron-phonon coupling

and thermal conductance at metal/non-metal interfaces," 2015 Materials Research Society Spring Meeting, San Francisco, CA, April 6 – 10, 2015 (Poster).

23) Scrymgeour, D. A., Ihlefeld, J., Foley, B.M., Michael, J., McKenzie, B., Hopkins, P.E., "Domain wall interface density control for tunable thermal conductivity," 2015 Materials Research Society Spring Meeting, San Francisco, CA, April 6 – 10, 2015.

Invited Presentations given by PI-Hopkins

1) "Engineering coherent transport, fractons, and Einstein oscillations in thermal conduction," 1st International Conference on Phononics and Thermal Energy Science (PTES2013), Shanghai, China, August 26 – September 4, 2013.

2) "Effects of temperature and anharmonicity at single interfaces: evidence from experiments," 2013 ASME International Mechanical Engineering Congress & Exposition, San Diego, CA, November 15 – 21, 2013 (Panel).

3) "Nanoscale heat transport: Fundamentals and current research," Sandia National Laboratories Professional Development Classes, Sandia National Laboratories, Albuquerque, NM, June 11, 2014 (4 hour tutorial).

4) "Pushing the limits and actively controlling the thermal conductivity of nanomaterials," 2014 Blavatnik Science Symposium, New York, NY, July 9, 2014.

5) "Controlling the thermal conductivity of ferroelectric nanostructures: Phonon interactions with domain boundaries," Center for Dielectrics and Piezoelectrics Fall Meeting, Raleigh, NC, November 3 – 4, 2014.

6) "Static and dynamic control of phonon transport in nanostructures: coherent interfaces and voltage-tunable thermal conductivity," Seminar, Department of Mechanical Engineering, *MIT*, Boston, MA, November 12, 2014.

7) "Static and dynamic control of phonon transport in ferroelectric oxide nanostructures: coherent interfaces and voltage-tunable thermal conductivity," 7th International Conference on Electroceramics (ICE2015), State College, PA, May 13 – 16, 2015.

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Principal Investigator Name

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Patrick E. Hopkins

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Abstract

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Archival Publications (published) during reporting period:

1) Q. Zhou, A. Cross, Y. Fu, A. Beling, B. Foley, P. Hopkins, and J. Campbell. Balanced InP/InGaAs photodiodes with 1.5-W output power. Photonics Journal, IEEE, 5(3):6800307, 2013.

2) P. E. Hopkins, C. Adamo, L. Ye, B. D. Huey, S. R. Lee, D. G. Schlom, and J. F. Ihlefeld. Effects of coherent ferroelastic domain walls on the thermal conductivity and kapitza conductance in bismuth ferrite. Applied Physics Letters, 102(12):121903, 2013.

3) R. E. Jones, J. C. Duda, X. W. Zhou, C. J. Kimmer, and P. E. Hopkins. Investigation of size and electronic effects on Kapitza conductance with non-equilibrium molecular dynamics. Applied Physics Letters, 102(18):183119, 2013.

4) C. B. Saltonstall, J. Serrano, P. M. Norris, P. E. Hopkins, and T. E. Beechem. Single element raman thermometry. Review of Scientific Instruments, 84(6):064903, 2013.

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6) A. Giri, B. M. Foley, and P. E. Hopkins. Influence of hot electron scattering and electron-phonon interactions on thermal boundary conductance at metal/nonmetal interfaces. Journal of Heat Transfer, 136:092401, 2014.

7) A. Giri, J. T. Gaskins, B. M. Foley, R. Cheaito, and P. E. Hopkins. Experimental evidence of excited electron number density and temperature effects on electron- phonon coupling in gold films. Journal of Applied Physics, 117(4):044305, 2015.

8) A. Giri, J. T. Gaskins, B. F. Donovan, C. Szwejkowski, R. J. Warzoha, M. A. Rodriguez, J. Ihlefeld, and P. E. Hopkins. Mechanisms of nonequilibrium electron- phonon coupling and thermal conductance at interfaces. Journal of Applied Physics, 117(10):105105, 2015.

9) R. Cheaito, K. Hattar, J. T. Gaskins, A. K. Yadav, J. C. Duda, T. E. Beechem, J. F. Ihlefeld, E. S. Piekos, J. K. Baldwin, A. Misra, and P. E. Hopkins. Thermal flux limited electron Kapitza conductance in copper-niobium multilayers. Applied Physics Letters, 106:093114, 2015.

10) J. F. Ihlefeld, B. M. Foley, D. A. Scrymgeour, J. R. Michael, B. B. McKenzie, D. L. Medlin, M. Wallace, S. Trolier-McKinstry, and P. E. Hopkins. Room-temperature voltage tunable phonon thermal conduc- tivity via reconfigurable interfaces in ferroelectric thin films. Nano Letters, 15:1791–1795, 2015.
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Research Objectives

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